Optics of Nanostructured Metallodielectrics

Miriam Deutsch, University of Oregon, DMR-02-39273

This project focuses on synthesis of poly-crystalline silver nano-shells on colloidal silica spheres (Fig. 1,) and characterization of the composite materials. White light extinction spectra reveal unique resonances, although the nanoshell is incomplete and highly granular, indicating the presence of coherent plasmon oscillations in the percolative nanoshells. (Fig. 2.) The resonances shift significantly when the refractive index of the surrounding medium is changed, demonstrating the sensing capabilities of these materials (Fig. 3.) When a single sphere is illuminated with ultrafast laser pulses, a strong frequency doubled signal, along with a broad super-continuum are observed (Fig. 4.), indicating large enhancements of nonlinear optical processes.

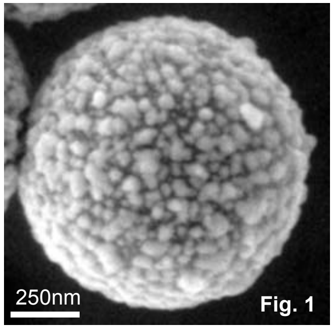

Fig. 1

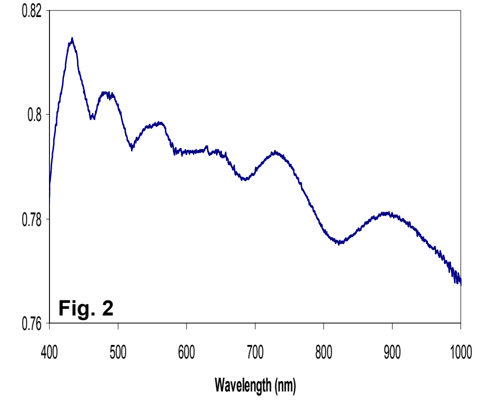

Fig. 2

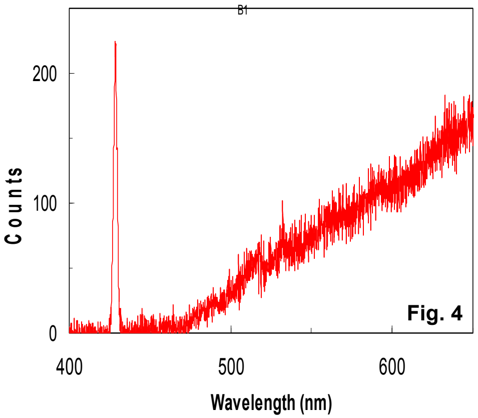

Fig. 4

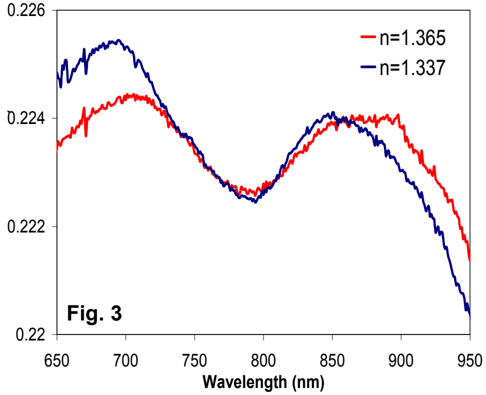

Fig. 3

The Research described here focuses on the synthesis and characterization of new optical materials. Materials periodically patterned on optical wavelength scales, such as three-dimensional photonic crystals hold great promise for generating and controlling optical signals. Examples include enhanced optical sensing capabilities of nano-structured materials, and nonlinear photonic crystals for optical logic applications. Both of these require detailed knowledge of synthesis of materials at the nano- and micron length scales, as well as understanding of the materials properties at these size scales.

Of particular interest to us is a new class of photonic materials, known as metallo-dielectric photonic structures. These materials are of immense interest due to the unusual dispersive effects present in metals, as well as greatly enhanced optical nonlinearities which have been demonstrated in these systems. These unique phenomena are mediated by the excitation of surface plasmon polaritons at the metal-dielectric interfaces. Such low-dimensional excitations are confined to dimensions much smaller than the wavelength of light, thus facilitating optical interactions at sub-diffraction-limit volumes. The rich optical phenomena offered by metallo-dielectrics therefore holds tremendous potential for novel optical materials, with unique spectral response and enhanced optical nonlinearities.

Our research focuses on fabricating self-assembled metallo-dielectric photonic crystals using novel colloidal building blocks, synthesized in our lab. These consist of silica sphere cores coated with highly granular silver nano-shells, as shown above. The coating process employs a known electroless deposition process, which we have modified to proceed slowly at low temperatures. The silver deposits directly on the spheres, without any chemical surface-functionalization of the silica (unlike in most other core-shell synthesis protocols, where surface-functional groups are required to ensure the growth of metal on the core particles.) This is advantageous when analyzing the optical properties of these materials, since the presence of organic surface-groups will alter the optical response at the metal interfaces, as well as prove detrimental under intense laser excitation. In our growth process shell thickness and morphology are controlled via concentration of the reactants, and temperature and duration of the reaction. This method allows us to grow porous (incomplete) nano-shells, similar to random percolative thin metal films.

We are currently studying the optical response of single isolated spheres, as it proves to strongly deviate from that of better-understood core-shell particles with smooth (complete) nano-shells. We have ascertained that the distinct plasmon resonances which we observe result from the unique nano-crystalline structure of the silver coatings. Modeling of linear light scattering as in Fig. 2 is under way, and a publication disseminating our findings is currently being prepared.

Single coated spheres also exhibit strong nonlinear optical response. We have observed both frequency doubling and white-light generation spanning the entire visible spectrum from an isolated sphere. Experimental studies and characterization of these phenomena are under way.

Two conference submissions detailing this work have been accepted for presentation at the MRS Fall 2004 Conference in Boston.

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Education:

One undergraduate (Daniel Cassell) and five graduate students (Jason Bouwman, Sarah Emmons, Charles Rohde, Aiqing Chen and Keisuke Hasegawa) are contributing to this work. Daniel Cassell has graduated from the University of Oregon in 2004 with a degree in Physics. Jason Bouwman was an NSF IGERT Fellow and has graduated with a Master's in Chemistry. Sarah Emmons is a Chemistry graduate student. She is the recipient of the NSF GK-12 fellowship. Charles Rhode is a Physics graduate student. He was an NSF IGERT fellow during the 2002-2003 academic year. Aiqing Chen and Keisuke Hasegawa are Physics graduate students.

Outreach:

The PI's research is interdisciplinary, and therefore draws interest both from the physics and chemistry communities.

The PI has been participating in the REU program.

The PI has participated in the Community Conversation Series at the University of Oregon. These include conversation events designed to stimulate intellectual activity among students and community members. The PI participated as panel speaker on the topic of Nanotechnology.